Reprinted from Optics Letters

Asynchronous optical sampling: a new combustion diagnostic for potential use in turbulent, high-pressure flames

R. J. Kneisler and F. E. Lytle

Department of Chemistry, Purdue University, West Lafayette, Indiana 47907

G. J. Fiechtner, Y. Jiang, G. B. King, and N. M. Laurendeau

Flame Diagnostics Laboratory, School of Mechanical Engineering, Purdue University, West Lafayette, Indiana 47907

Received June 24, 1988; accepted December 27, 1988

Asynchronous optical sampling (ASOPS) is a pump–probe method that has strong potential for use in turbulent, high-pressure flames. We show that rapid measurement of species number density can be achieved by maintaining a constant beat frequency between the mode-locking frequencies of the pump and probe lasers. We also describe the instrumental timing parameters for ASOPS and consider the optimization of these parameters. Measurement of the nanosecond decay for electronically excited sodium in an atmospheric flame demonstrates the viability of the ASOPS technique in highly quenched flame environments.

Absolute number density measurements in flames are difficult to derive from laser-induced fluorescence signals because the rate of fluorescence quenching due to collisional deexcitation is not well known.[1] Laser-saturated fluorescence can be used to avoid the quenching dependence,[2] but this technique is limited by the number of molecules that can be effectively saturated. Thus in flames above 1 atm the measurement of number density requires that picosecond lasers be used to determine directly the rapid quenching rates. Previous investigators have used picosecond lasers to do time-resolved laser-induced fluorescence experiments on OH at atmospheric pressure using either a streak camera[3,4] or a time-correlated single-photon counting scheme.[5] Although these experiments have the temporal resolution necessary to obtain the needed quenching rates, the amount of time necessary to obtain the data is much greater than the time scale of turbulence.

In this Letter we present a new technique for potential use as a combustion diagnostic in turbulent, high-pressure flames. The technique, asynchronous optical sampling (ASOPS), is a pump–probe method, but it overcomes many of the temporal problems inherent in that method.[6,7] Furthermore, corrections for the effects of quenching can be obtained on a time scale necessary for practical combustion measurements.[8,9] In the ASOPS method, rather than using a single Nd:YAG laser to construct both the pump and probe beams and an optical delay line to control the relative timing between the two pulses, two Nd:YAG lasers are used to generate the pump and probe beams separately. Central to the method is the fact that the two Nd:YAG lasers are mode locked at slightly different frequencies. The mode-locking frequencies of these two lasers are carefully controlled to maintain a constant beat frequency, which creates a periodic relative phase walk out between the pump and the probe lasers. This has the same effect as varying the optical delay line in the conventional pump–probe method; however, the time needed to observe the population decay of an excited energy level can be reduced from several minutes to less than a millisecond.

The process is illustrated in Fig. 1(a), which shows the excited-state population produced by several pump pulses and the temporal position of several probe pulses. Each successive probe pulse is delayed in time (relative to the pump-pulse train) by a constantly increasing duration that is determined by the beat frequency of the system. Thus each probe pulse samples the excited-state population at a slightly later time than the preceding probe pulse. The sampling process repeats itself when the cumulative delay equals the period of the pump laser. Therefore in contrast to a conventional pump–probe instrument, there is no need to modulate the amplitude of either beam to employ synchronous detection.

Figure 1(b) illustrates the change in probe intensity that occurs on stimulated emission from the excited-state population shown in Fig. 1(a). The net effect of the ASOPS technique is that a small-amplitude waveform, which is directly related to the fluorescence decay of the species under study, is impressed onto the

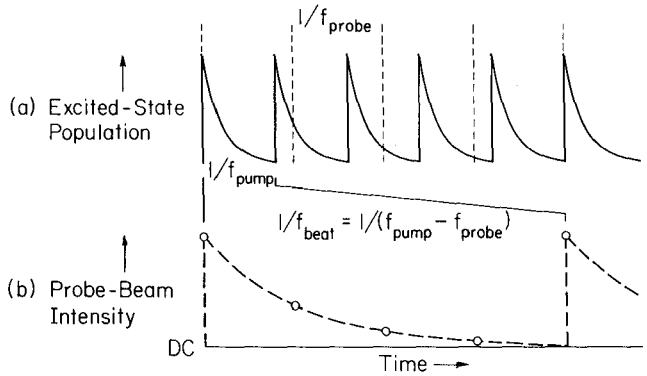

Fig. 1. ASOPS timing diagram showing (a) the excited-state population and (b) the probe-beam intensity. The probe pulses are indicated by the vertical dashed lines.

0146-9592/89/050260-03$2.00/0

Table 1. Operating Parameters for Various Beat Frequencies

	Beat Frequency		
	10 kHz	100 kHz	1 MHz
Repetition rate (f_{pump}) (MHz)	~82	~82	~246
Free temporal range ($1/f_{pump}$) (nsec)	12.2	12.2	4.1
Collection time ($1/f_{beat}$) (μsec)	100	10	1
Samples per decay (f_{probe}/f_{beat})	8200	820	246
Sampling interval ($f_{beat}/f_{probe}f_{pump}$) (psec)	1.5	15	17
Duty cycle (for τ = 1 nsec) (%)	25	25	74

probe-laser intensity. In essence, a temporal transformation of the excited-state decay is performed with the time scaled by the factor $[f_{pump}/(f_{pump} - f_{probe})]$, where f is the repetition rate of the two lasers. The ASOPS technique is thus an optical analogue of the sampling oscilloscope.

The beat frequency of the ASOPS instrument is determined by the difference in laser repetition rates, or $f_{beat} = f_{pump} - f_{probe}$. The period of the pump laser, $1/f_{pump}$, determines the free temporal range, which is the maximum time available for a single decay of the excited-state population. The inverse of the beat frequency, $1/f_{beat}$, represents the collection time for a single decay. The total number of points sampled during the decay is determined by f_{probe}/f_{beat}. The temporal difference between each sampled point within the decay profile is the sampling interval. The sampling interval and thus the temporal resolution is determined by the difference in laser periods, i.e., $f_{beat}/(f_{probe}f_{pump})$. If we assume that the signal is zero after 3τ, where τ is the excited-state lifetime, then the percent duty cycle is given by $100(3\tau f_{pump})$.

Table 1 gives the above operating parameters for various beat frequencies and laser repetition rates. Several factors must be taken into account to determine the optimum ASOPS parameters. First, to make measurements on the time scale of turbulence it is necessary to collect enough data for adequate signal averaging in ~1 msec. This criterion indicates that we should use the largest possible beat frequency. However, the beat frequency is limited by the physical cavity length of the lasers and the natural acoustic resonance frequencies of the mode-locking prisms. In addition, since the ASOPS signal is carried on the probe beam, the beat frequency must be chosen so as not to coincide with any noise in the frequency spectrum of the laser. The second factor important to system optimization concerns the sampling interval, which must be small enough so that the ASOPS measurement has sufficient temporal resolution to reconstruct the excited-state decay. This criterion sets a natural upper limit to the beat frequency because the sampling interval is proportional to f_{beat}. Third, the duty cycle should be as close to 100% as possible. As previously shown, the duty cycle increases with the pump-laser repetition rate and is independent of the beat frequency. The pump-laser repetition rate can be increased by a technique called third-harmonic mode locking. As can be seen from Table 1, this approach would provide the ideal case for ASOPS.

Since the ASOPS technique requires that the pump and probe lasers operate at slightly different repetition rates, the instrument must be constructed from two independent mode-locked laser systems.[7] The mode-locking frequencies are generated by two frequency synthesizers (accurate to 0.1 Hz) operated in a master–slave (i.e., phase-locked) configuration to minimize drift in the beat frequency of the system. Both the pump and probe beams consist of an ~82-MHz train of ~5-psec pulses, tunable (using Rhodamine 6G) from 560 to 640 nm. The measured impulse response of the system, as determined by a cross-correlation method described previously,[7] is ~7 psec; this compares favorably to the convolution of the pump and the probe pulses, verifying the stability of the

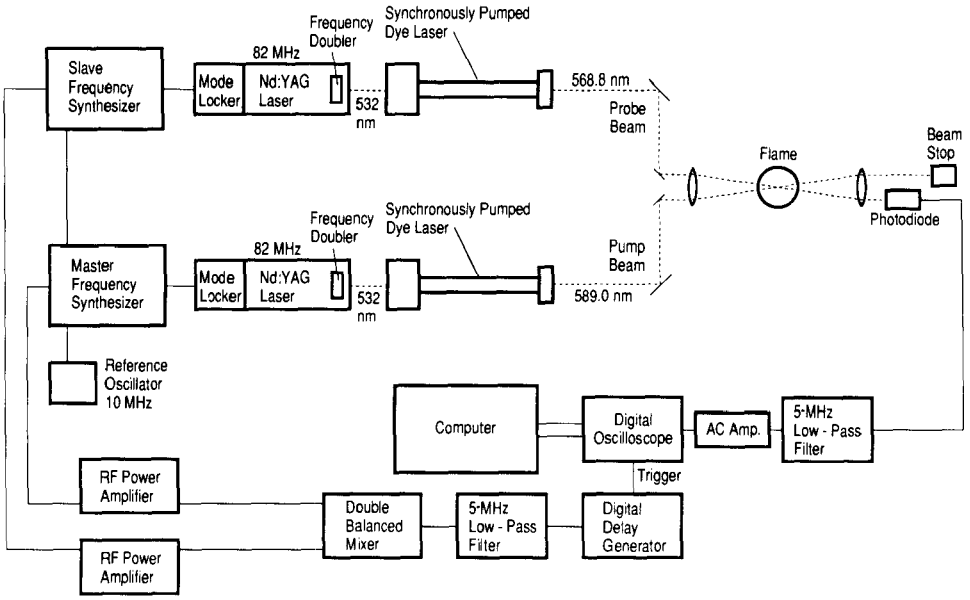

Fig. 2. Block diagram of the basic ASOPS instrument.

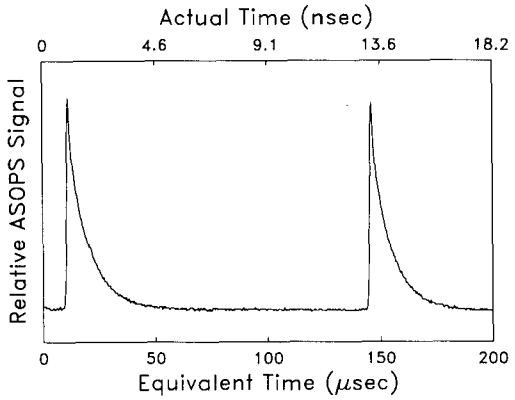

Fig. 3. ASOPS signal for atomic sodium in a 1-atm flame. The equivalent decay time is 13.2 μsec, which corresponds to an actual decay time of 1.2 nsec.

master–slave electronics scheme. Three-plate Lyot (birefringent) filters placed in the dye-laser cavities result in nearly transform-limited bandwidths of 120 GHz (Ref. 10) ($\Delta\lambda$ = 0.14 nm) for both the pump and probe lasers. Throughout the center of the tuning range average powers in excess of 120 mW are obtained by using 800–900 mW of pumping power.

A block diagram of the basic ASOPS instrument is shown in Fig. 2. To obtain the trigger signal a synchronous voltage output from each synthesizer is first amplified with an rf power amplifier (Electronic Navigation Industries 503L) and then electronically mixed by a double-balanced mixer (Anzac MDC-161). Note that the output frequency of the synthesizer is half that of the optical repetition rate; electronic frequency doublers will eventually be used to generate a trigger signal at the beat frequency of the system. The mixer output passes through a 4LM5-3-CD Texscan 5-MHz low-pass filter to remove any high frequencies leaking through the mixer. The output of the filter then enters a digital delay generator (Berkeley Nucleonics Corporation 7095), which produces a stable 0–5-V trigger pulse of variable width. Unlike the optical triggering scheme previously employed,[7,11] this new electronic scheme offers much less temporal jitter in the trigger pulse and thus an improved signal-to-noise ratio.

The pump and the probe beams are first passed through calibrated neutral-density filters, which provide a convenient means of varying the power in either beam. The two beams then pass through a single focusing lens (f = 200 mm) and cross in the flame. The probe beam is recollimated by a matching lens and monitored by a photodiode (EG&G SGD-100A), the output of which is filtered with a 4LM5-3-CD Texscan 5-MHz low-pass filter to remove the high-frequency noise. The output from the filter is amplified by a C-COR 4375-A wideband ac amplifier. The output from the amplifier is then directed to a digitizing oscilloscope (Hewlett-Packard 54100A) that is triggered at half the beat frequency of the system.

Currently the ASOPS repetition rates are f_{pump} = 81.5929242 MHz and f_{probe} = 81.5855038 MHz, giving a beat frequency f_{beat} = 7.4204 kHz. The pump power is 65 mW at λ = 589.0 nm, and the probe power is 2.5 mW at λ = 568.8 nm. The beam diameters are 50 μm, and the interaction length in the flame is 2.3 mm. The probe-beam signal is amplified by a factor of 100.

As the first substantive test of ASOPS in a combustion environment, we demonstrate the detection of sodium in an atomic-absorption slot burner using a $CH_4/O_2/N_2$ mixture and a NaCl solution fed with a multistaltic pump. Figure 3 shows the ASOPS signal for sodium in an atmospheric flame. The pump beam is set to the sodium D_2 ($3S_{1/2} \rightarrow 3P_{3/2}$) transition (589.0 nm), while the probe beam is set to the $3P_{3/2} \rightarrow 4D_{5/2,3/2}$ transition (568.8 nm). Thus in this case the ASOPS signal is monitoring the loss in the probe-beam intensity. The equivalent exponential decay time of 13.2 μsec, which corresponds to an actual decay time of 1.2 nsec, indicates the total removal rate from the $3P_{3/2}$ state.

The decay curve in Fig. 3 was obtained by averaging over 1024 separate single-decay curves. Ideally the averaging process should take 146 msec to complete; however, because of the slow rearming rate of the oscilloscope, the averaging process took 39 sec. If the digital oscilloscope is replaced with a transient digitizer, sampling can eventually occur at the probe-laser repetition rate. The peak signal-to-noise ratio is 74. Suitable upgrades are possible with respect to the performance of both the Nd:YAG laser systems and the detection electronics. We estimate an ultimate ASOPS detection limit for sodium of 10^2 atoms/cm^3.

This research was sponsored by the U.S. Air Force Office of Scientific Research, Air Force Systems Command, USAF, under grant AFOSR-84-0323.

Y. Jiang is a visiting professor from the Department of Precision Instruments, Tsinghua University, Beijing, China.

References

1. P. W. Fairchild, G. P. Smith, and D. R. Crosley, J. Chem. Phys. **79,** 1975 (1983).
2. R. P. Lucht, D. W. Sweeney, and N. M. Laurendeau, Combust. Flame **50,** 189 (1983).
3. N. S. Bergano, P. A. Jaanimagi, M. M. Salour, and J. H. Bechtel, Opt. Lett. **8,** 443 (1983).
4. R. Schwarzwald, P. Monkhouse, and J. Wolfrum, Chem. Phys. Lett. **142,** 15 (1987).
5. Y. Takubo, T. Okamoto, N. Komine, and M. Yamamoto, Jpn. J. Appl. Phys. **26,** 416 (1987).
6. P. A. Elzinga, F. E. Lytle, Y. Jiang, G. B. King, and N. M. Laurendeau, Appl. Spectrosc. **41,** 2 (1987).
7. P. A. Elzinga, R. J. Kneisler, F. E. Lytle, Y. Jiang, G. B. King, and N. M. Laurendeau, Appl. Opt. **26,** 4303 (1987).
8. D. Stepowski and M. J. Cottereau, Appl. Opt. **18,** 354 (1979).
9. D. Stepowski and M. J. Cottereau, Combust. Flame **40,** 65 (1981).
10. T. Gray, Spectra-Physics Corporation, 1250 West Middlefield Road, Mountain View, California 94039 (personal communication, 1987).
11. G. J. Fiechtner, Y. Jiang, G. B. King, N. M. Laurendeau, R. J. Kneisler, and F. E. Lytle, "Determination of relative number density and decay rate for atomic sodium in an atmospheric premixed flame by asynchronous optical sampling," in *Twenty-Second International Symposium on Combustion* (The Combustion Institute, Pittsburgh, Pa., to be published).